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Optogalvanic transitions in lutetium in the 570–630 nm region

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Abstract : Laser optogalvanic (LOG) technique is now a well-established spectroscopic tool to study the spectra of atoms and molecules. Rare earth atoms have high melting point and normally their excitation is difficult using ordinary methods. The emission spectrum of the lutetium/neon hollow cathode lamp was recorded in the 500–750 nm region using a 0.5 m monochromator, a cooled photomultiplier coupled to a picoammeter and a chart recorder. The LOG spectrum of Lu/Ne lamp was recorded using an argon ion (Ar+) laser and a tunable linear dye laser with rhodamine 6G in the 570–630 nm region. The dye laser output power at its maximum was ~ 200 mW with nearly 40 GHz bandwidth. The lutetium lines wavelengths are expected to be accurate to within 1 nm. In the LOG spectrum, only 47 lines could be observed and some of these lines have already been observed by earlier workers. Three new lines have been identified here, whereas in the emission spectrum, in the same region, only 16 lines of neutral and singly-ionized lutetium besides neon lines were recorded. Three new lines are at 600.15 nm, 611.527 nm, 614.73 nm which are assigned to different transitions.

Keywords : LOG-laser optogalvanic technique, lutetium.

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1. Introduction

Spectral studies of rare earth atoms are rather scanty in the literature primarily because of their close-lying multiplet states resulting in a very complex structure. In most of the cases, they have high melting point and normally their excitation is difficult using ordinary methods.

Lutetium element has a closed '*f*' shell and has only two isotopes and it exhibits a relatively simpler spectrum, which has encouraged several workers [1–6] to study its spectrum using different techniques. Rare earth atoms often have several isotopes with significant abundance and with nuclear spin creating further congestion in the spectrum due to isotope effect and hyperfine splitting.

Zimmerman *et al* [7] reported the isotopic shift in the 542.2 nm line of lutetium. They have also studied the hyperfine structure of the transition $5d6s\ ^{22}D_{3/2} \rightarrow 5d6s6p\ ^{4}F_{3/2}$ (573.655 nm) of the two isotopes ^{175}Lu and ^{176}Lu. Nunneman *et al* [8] later on, investigated the hyperfine structure and isotopic shift in the 605.5 nm line of ^{176}Lu.

Keller *et al* [9] studied the hyperfine structure of the lutetium transition $5d6s6p\ ^{2}D_{3/2} \leftarrow 5d6s\ ^{22}D_{3/2}$ at 451.0 nm, both by Fourier transform technique in emission as well as by LOG spectroscopy. They concluded that the optogalvanic (OG) spectra showed a large deviation in relative intensities of the line from the intensities in the classically recorded spectrum. The relative intensity in the OG spectrum also changes when the laser power is increased. They have also measured the isotopic shift between ^{175}Lu and ^{176}Lu. Recently, Rao *et al* [10] recorded the LOG spectrum in ^{175}Lu using a single frequency dye laser and studied the hyperfine structure in ^{175}Lu. They reported the hyperfine splitting in the lower and the upper levels of many transitions of ^{175}Lu.

This analysis leads to several quantities such as hyperfine splitting constants *A* and *B*, nuclear spin, nuclear moment, isotope shift *etc*. These workers also observed eight new lutetium lines at 566.302 nm, 579.32 nm, 583.0 nm, 584.25 nm, 601.98 nm, 605.07 nm, 605.15 nm and 608.42 nm. Lutetium has two stable natural

isotopes ^{175}Lu (97.4%) and ^{176}Lu (2.6%). Since ^{176}Lu has a very low natural abundance, the signals relating to this isotope will appear very weak if found at all. The optogalvanic as well as emission spectra of Lu have been investigated for two reasons, namely, (a) in the 570–630 nm region, only 8 lines of Lu are reported in earlier literature while more lines are expected [6], (b) LOG technique is very sensitive so that other lines may be observed using this technique.

2. Experimental

A commercial lutetium hollow-cathode lamp with neon as a buffer gas has been the sample source in our experiments. The hollow cathode lamp (Lu/Neon) was obtained from a commercial manufacturer (Hamamatsu, Japan) who is known to prepare 'elements' with high purity. The chances of an impurity is thus eliminated from the signal obtained. DC voltage from a well-stabilized DC power supply has been used to excite the discharge with a maximum current of ~8 mA at around 235 volts d.c. As a first step, we have recorded the emission spectrum of the lamp in the region 5000–7000 Å using a 0.5 m monochromator. A cooled photomultiplier tube coupled to a picoammeter and a chart recorder, has been used for detection of emission. The use of a hollow cathode lamp at slightly higher current gives appreciably larger concentration of the electrode material in the discharge due to sputtering, thereby increasing the chance of detecting additional lines. The experimental set-up used for detection of Lu/Neon lines is shown in Figure 1.

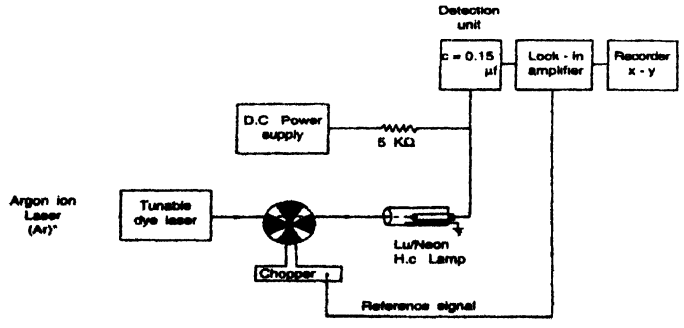

Figure 1. Optogalvanic experimental set-up for lutetium.

We have also recorded the LOG spectrum of the Lu-Ne lamp. The laser used in our study is a tunable linear dye laser with Rhodamine 6G dye pumped by an Ar+ laser. The dye laser output was focussed into the hollow cathode lamp, through a 20 cm focal length convergent lens. The beam was chopped at 144 Hz using a Precision Instrument mechanical chopper. The dye laser output power at its maximum was ~200 mW with nearly 4(GHz bandwidth. The dye laser was tuned using a thre plate birefringence filter. The discharge condition was th same as while recording the emission spectrum. A ballas resistor of 5 KΩ resistance and a capacitor of 0.15 μF capacity were used in the detection circuit for separatin the DC signal.

The signal was optimized by varying the variou experimental parameters. The OG signals due to Lu line appeared embedded in the more abundant neon spectrun and one had to adjust the sensitivity and time constant o the lock-in-amplifier to see the former. The input signa to a lock-in-amplifier is usually a time-varying sinal signal oscillating at the frequency of a mechanical choppe used for providing a modulated incident optical beam The input signal is usually amplified at the choppin frequency before it is synchronously demodulated i order to detect the signal. Amplification is carried ou without adding too much of 'noise' and thus helps i securing a reasonable signal to noise ratio.

The key element of the lock-in-amplifier is the mixe which operates a phase-sensitive switch. When a signa reaches a mixer, its appearance is nearly sinusoidal depending upon the type of pre-detection filtering used For the efficient use of mixer circuits, these must b capable of withstanding signal so that nonlinear respons does not take place without overloading (we have used EG & G model 186A and SR530 Stanford Researc Systems, USA) lock-in-amplifiers in our studies. Thes models have the capability to detect a signal as low a few millivolts and can cope with reference frequency o several KHz. The time constant of 0.1 second wa chosen after recording several other time constant value like 1 second, where only neon lines appeared.

The measurements were repeated by varying th discharge voltage, the resistance of the filtering circui and the time constant of the detector. It has been found that if the time constant is equal to or greater than 1.(second, only neon lines are seen. However, if the time constant is reduced below 1.0 second, some lutetium lines start to appear with weak intensity. The final recording of the spectrum was made with a time-constant of 0.1 second (which is the lowest in this case). Since the Lu lines are much weaker compared to neon lines, we had to saturate the OG signal for neon lines for recording Lu lines with appreciable intensity. The signals were recorded with the help of a Digital Electronics Omniscribe double pen *xy* recorder. The strong neon lines observed were used for calibration of the

wavelengths of the Lu lines. The wavelengths of Lu lines were expected to be accurate to within 1 nm.

3. Results

In the emission spectrum between 570–630 nm, we obtained 72 lines belonging to neutral neon (Ne I) and lutetium (Lu I). We could also notice some lines due to singly ionized lutetium (Lu II).

In the laser optogalvanic spectrum, only 47 lines could be observed which contain three new lines of Lu besides the lines observed by earlier workers [10]. In the emission spectrum in the same region, we have obtained only 16 lines of neutral and singly ionized lutetium (Lu II) besides neon lines.

In the LOG experiment, the observation of lutetium lines was found to be difficult. They appear only when the time constant of the lock-in-amplifier was adjusted to be ~0.1 sec. Any increase in the time constant reduces the intensity of the LOG signal associated with Lu lines. Similarly, if the current was reduced below 18 mA, the intensity of the Lu lines decreases rapidly and only neon signals were seen. A part of the spectrum is shown in Figure 2. The optogalvanic signals appear both positive and negative in the recording. All the signals observed in the LOG spectrum are due to neutral Lu. Table 1 gives the observed data of emission and LOG lines of lutetium/

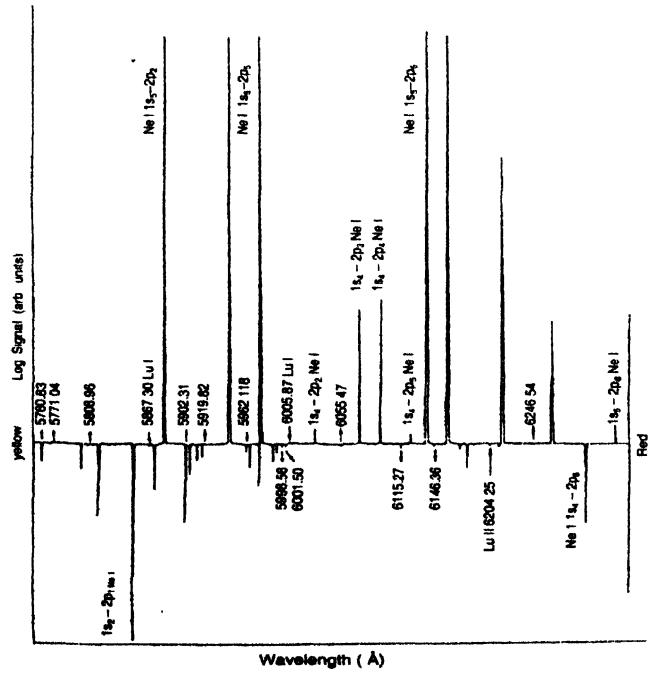

Figure 2. Laser optogalvanic spectrum of lutetium/neon.

Table 1. Emission lines and optogalvanic lines of lutetium/neon in the 5700 Å - 6300 Å region.

Sl. No.	LOG lines (Å) of Lu/neon	Emission lines of lutetium (Å)	Reported lines of NSRDS – data of lutetium (Å)
1.	–	5736.55 (Lu I)	5736.55 (Lu I)
2.	5760.83	–	5748.71 (Lu III)
3	5771.04	5775 40 (Lu I)	5775.40 (Lu I)
4.	5800.21	5800.59 (Lu I)	5800.59 (Lu I)
5.	5808 96	-	–
6.	5817 72	–	–
		5830.00 (Lu I)	
7	5851.26	–	5860.79 (Lu I)
8.	5867.31	5866.30 (Lu I)	5866.30 (Lu I)
9	5881.89	-	–
10.	5902.32	–	–
11.	5906 69	–	–
12.	5913.98	–	–
13.	5919.82	–	–
14.	5934 40	–	–
15.	5946.07	–	–
16.	5962.12	–	–
17.	5966.49	–	–
18.	5975 24	-	–
19.	5976.70	–	–
20.	5989.83	5983.90 (Lu I)	5983.90 (Lu II)
21.	5992.75	-	–
22.	5998.58	5997.13 (Lu I)	5997.13 (Lu I)
23.	6001.50	-	–
24.	6005.87	6004.52 (Lu I)	6004.52 (Lu I)
25.	6030.67	–	–
26.	6042.34	6041.66 (Lu I)	6041.66 (Lu I)
27.	6055.46	6055.03 (Lu I)	6055.03 (Lu I)
28.	6065.68	–	–
29.	6075.88	–	–
30.	-	6084.17 (Lu I)	–
31.	6096.31	–	–
32	6115.27	–	–
33.	6126.94	–	–
34.	6138.61	–	–
35.	6141.52	6140.71 (Lu I)	6140.71 (Lu I)
36.	6147.36	–	–
37.	–	–	6159.94 (Lu II)
38.	6161.40	–	–
39.	6173.61	–	–
40.	6180.91	–	–
41.	6186.74	–	–
			6197.96 (Lu III)

Table 1. *(Cont'd.)*

42.	6204.24	–	6198.13 (Lu III)
43.	6217.99	–	6199.66 (Lu II)
44.	6217.37	6221.87 (Lu I)	6221 87 (Lu II)
45.	–	–	6228.87 (Lu II)
46.	.	.	6235.36 (Lu II)
47.	–	–	6242.34 (Lu II)
48.	6246.54	6248.80 (Lu II)	6248.80 (Lu II)
49.	6266.96	–	-
50.	6300.57	–	-
51.	6300.57	–	-
52.	6329.28	–	-

neon in the region of 570 nm – 630 nm. The error in the observation is of the order of 1 nm [11]. The lines that match between emission and the observed LOG spectra are 573.65 nm, 577.54 nm, 579.325 nm, 580.059 nm, 583.0 nm, 586.630 nm, 598.39 nm, 599.713 nm, 600.452 nm, 604.166 nm, 605.503 nm, 608.417 nm, 614.071 nm, 622.187 nm, 623.536 nm 624.88 nm.

Three new lines are shown in Figure 2 which have been observed at 600.15 nm, 611.527 nm and 614.736 nm, respectively.

4. Conclusions

(i) In the emission spectrum we have obtained 72 lines of neutral neon, neutral lutetium (Lu I), singly ionized lutetium (Lu II), and doubly ionized lutetium in the 570–630 nm region.

(ii) Three new lines in the laser optogalvanic spectrum are observed by us and their wavelengths are 600.15 nm, 611.527 nm and 614.736 nm which are assigned to different transitions.

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